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Carbon dioxide emission reduction using molten carbonate fuel cell systems

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A B S T R A C T

The contribution of the molten carbonate fuel cell system (MCFCs) to carbon dioxide (CO_2) emission reduction in power application is analyzed. MCFCs can separate and concentrate CO_2 emitted from traditional thermal power plants (PPs) without reducing the plant's overall energy efficiency. MCFCs can also be used by itself as an effective CO_2 separator or concentrator by managing the anode gas stream to increase the heat utilization of the system. The CO_2 separated and concentrated by MCFCs is most effectively captured by condensation. MCFCs is currently used as a CO_2 separator only to a limited extent due to its high cost and relatively small scale operation. However, MCFCs will substantially contribute to reduce CO_2 emissions in power generation applications in the near future.
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Contents

1. Introduction

Climate change is believed to be one of the most important issues in this century. Carbon dioxide (CO_2) emissions should be reduced to prevent global warming. Although renewable and sustainable energy can significantly contribute to reduce CO_2 emissions, it will take a very long time for them to be easily exploited and widely used. Therefore, fossil fuels will continue to be used because they are relatively abundant and economic. However, because they necessarily involve great CO_2 emissions, CO_2 capture and storage (or utilization) technology should be developed and employed to mitigate CO_2 emissions.

Separation using membranes, chemical looping and molten carbonate fuel cell system (MCFCs) has been considered to utilize advanced CO_2 capture technologies because they can effectively

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separate and relatively easily concentrate CO_2 emitted in the processes [1,2]. Therefore, they are believed to be one of the most promising options to reduce CO_2 emission. However, there are many challenges to be addressed in each technology. Regarding membrane technology, many selective membranes for CO_2 capture have been developed such as polymeric, mixed ionic–electronic (solid electrolyte–metal) films as well as metallic (palladium)-based materials [2,3–11]. However, the technology is subject to the limitations of high operation pressure, CO_2 concentration and process scale-up [9,10,12].

Chemical looping combustion has the great potential to separate CO_2 from flue gas. Many papers [13–23] have reported the efficiency of various metal oxide catalysts and their regeneration performance. Despite their promising results, further study on more reactive and stable catalysts, which can be regenerated for numerous cycles, is still required. Catalyst attrition and deactivation due to carbon deposition are another primary issue that needs to be addressed for practical use [24,25]. Although these two technologies may be successfully developed in the near future, they may be basically less competitive with MCFCs because the original purpose of membrane and chemical looping is solely to separate CO_2 in gas mixture, while the CO_2 separation potential of MCFCs is its surplus technology. MCFCs has particularly many technologically advantageous features for CO_2 capture [26–40]. Firstly, MCFCs is operated with a high electrical efficiency due to the absence of any mixing of air and fuel in the process. Secondly, CO_2 is used as a reactant in the cathode at a constant concentration and flow-rate during the operation. Finally, a MCFC-hybrid

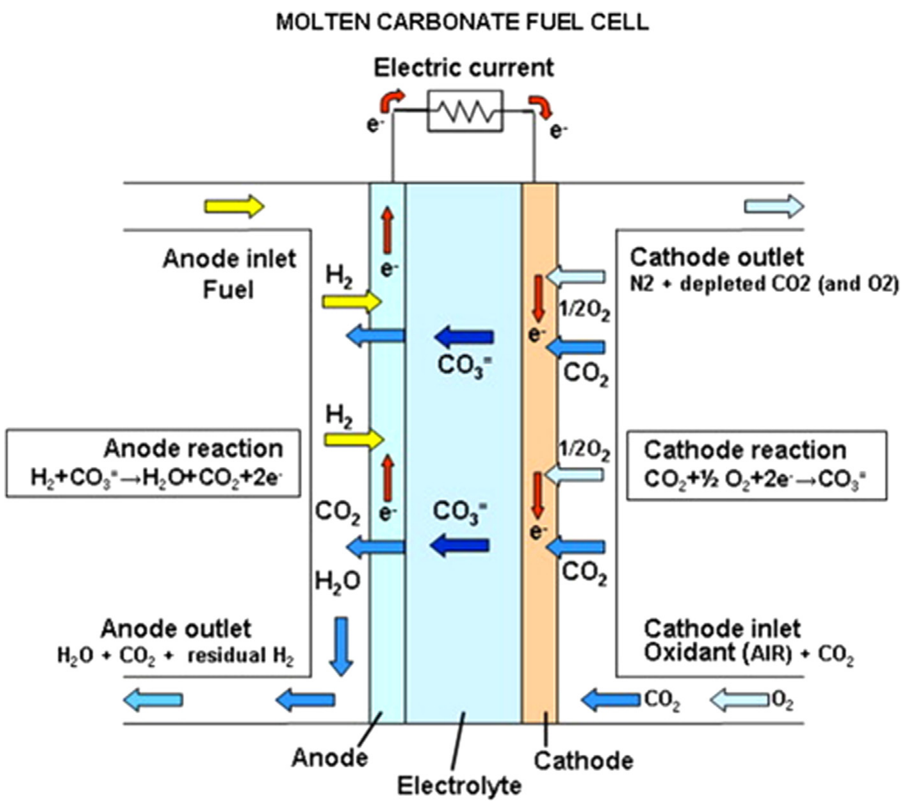


Fig. 1. Working scheme of MCFC [38].

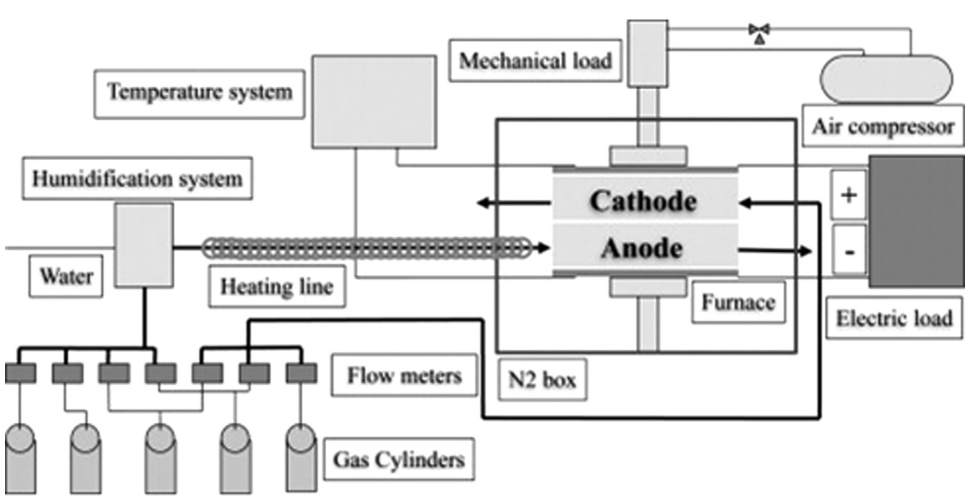


Fig. 2. Flowsheet of the experimental setup of the test bench [33].

system, which integrates MCFCs with the tradition thermal power plant (PP), can be effectively operated with great energy efficiency and possibly large CO_2 emission reduction.

CO_2 separation technology using MCFCs may have been significantly influenced from the literature [41–44] that investigated the CO_2 separation performance of the ceramic membrane cell (CMC) in which an alkali carbonate substance was used as the electrolyte. Electric current is supplied to the CMC for CO_2 separation and transportation. Despite the electricity consumption in the system, the CMC can continuously and in-situ separate highly concentrated CO_2 from the specific emission sources without any solvent [45]. In MCFCs, however, the fuel is supplied to an anode and the same reaction of CMC with CO_2 as the reactant is carried out to generate electricity as shown in Fig. 1.

Although MCFCs coupled with CMC was recently introduced [46], CO_2 can be separated and concentrated by MCFCs for capture.

In fact, Chugoku Electric Power Co. in Japan has demonstrated a CO_2 separation and concentration process employing MCFCs (10 kW) in coal-fired PP (CFPP; 1000 MW) in 2006. According to their report [47], the system could concentrate a CO_2 composition to 80% from 13% in CFPP exhaust gas.

In addition, in 2012, Discepoli et al. [33] conducted experimental tests to evaluate the potential of MCFCs as CO_2 concentrators using single MCFCs with an effective cell area of 55 cm^2 as shown in Fig. 2.

The gas mixture including CO_2 was prepared with the same composition of real NGCC exhaust gases as used in cathode gas. The final result from the experiment and modeling was that the CO_2 capture ratio of the MCFCs was strongly dependent on the partial pressure ratio of O_2 and CO_2 (P_{O2}/P_{CO2}) in the cathode and it maximized to 70% at CO_2 concentration over 8%.

Research on MCFCs has been conducted for more than 35 years and has rapidly developed its technology leading to an overall efficiency of about 75% (combined heat and power generation (CHP) based). Recently, MCFCs has begun to be commercialized and a total capacity of 53 MW has already been installed and operated in 20 sites throughout South Korea [48,49]. These commercial operations have received incentives from FIT and have been accepted as certified new energy under the RPS regulations. In addition, the demand of MCFCs is expected to increase according to increasing CH_4 production (shale gas) worldwide. In addition, the issue of climate change due to CO_2 emission has received great attention. Therefore, it may be constructive to review the quantitative contribution of the MCFCs to reduce CO_2 emissions in power generation application fields. The present paper analyzes MCFCs as a CO_2 separator and concentrator based on the latest research works. This paper presents a technology assessment, including the system layout, performance and CO_2 capture ratio, as well as the challenges.

2. Contribution of MCFCs to CO_2 emission reduction

In natural gas (NG)-based thermal PPs such as NG combined cycle (NGCC), NGPP (or integrated gasification (IG)CC), and MCFC, their contribution to reducing CO_2 emissions can be basically determined by comparing their energy efficiency (kW h/kg CH_4) and CO_2 emission coefficient (CEC) (kg CO_2/kW h). Although these two values may have a similar meaning, because they have not always exhibited a directly proportional relation in every system, they are separately explained in this paper.

The analysis of the plant configuration is also very useful to compare its contribution to CO_2 emission reduction because it is a very important systematic factor which can influence the CO_2 capture ratio. Therefore, a detailed investigation of the energy efficiency, CEC and system configuration of the MCFCs is presented in comparison with other NG-based PPs.

2.1. High energy efficiency

The power efficiency and CEC of the traditional thermal PPs, including commercialized MCFC, are listed in Table 1 [35,50–57].

The power efficiency of the stand-alone MCFCs [48] is higher than that of traditional NGPP and NGCC, but less than that of state-of-the-art, advanced NGCC, which has a claimed efficiency (LHV based) of approximately 60% [57]. However, considering the heat recovery from the system, the overall efficiency of MCFCs is possibly increased to approximately 60–90% [48,58] while CEC is decreased to under 236 kg/MW h. This is the lowest value among the CEC of the PP. In addition, biogas has been practically used as fuel for the MCFCs cogeneration system producing pure H_2 with high energy efficiency [59]. Therefore, MCFCs has more potential to reduce CO_2 emissions compared to other PPs due to its relatively high energy efficiency, fuel diversity and cogeneration ability. In addition, although the status of the MCFC-gas turbine (GT) hybrid system technology is almost at the conceptual level or partially demonstration stage, many papers [60–68] have reported that the system may be operated with even higher efficiency in the future. However, many problems and challenges should be addressed for its practical use. Because there are significantly different operational features and unlikely dynamic response between MCFCs and GT system, an innovative technology such as new GT design and its connection method to MCFC, which facilitates the effective operation and control of the hybrid system, should be developed firstly developed. In addition, the scale-up technology of the system is also very crucial for its commercialization. Many papers [69–75] have reported that if these are successfully solved, the efficiency of the MCFC–GT hybrid system will exceed 70% and the CO_2 emissions will be substantially reduced.

Table 1
Power efficiency and CEC of traditional thermal power plants (PPs).

Power plants	Power efficiency (%)	CO_2 emission coefficient (kg/MW h)
CFPP[a]	35.9–41.5	736–1290 [60,52]
NGPP	35.9–40.9	514–713 [53]
NGCC	40.6–58.7 (> 60)[b]	344–391 [35,36,53,56,57]
Oil thermal	31.8–35.2	689–890 [52]
MCFC	~47	445[c], 236–309[d] [48]

[a] Including advanced pulverized and pressurized fluidized bed CFPP.
[b] NGCC using H-Class GT [57].
[c] Without heat recovery.
[d] With heat recovery.

Table 2
Exhaust gas compositions of thermal power plants (PPs).

Power plants			Exhaust gas compositions (mol%)						
			H_2	CH_4	CO_2	CO	N_2	O_2	Water
CFPP [50]					15		78.5	6	0.5
NGPP [76]					8		73	3	16
NGCC [76,77]					3–4		76	13–14	7
MCFC	Overall	[48]			5		65	10	20
		[78]			9.3		77.5	2.3	10.9
	Anode	[79][a]	17.3	0	31.1	9			42.6
		[80][b]	6.1	0	48.7	5.2			39.4
		[81]	8.4	2.4	40.1	5.3			43.8
	Cathode	[81]			5.8		79.3	9.5	5.4
		[82]			7.9		81.8	6.6	3.7

[a] At fuel utilization of 60.82%.
[b] At fuel utilization of 78.5% and power efficiency of 50%.

2.2. CO_2 separator and concentrator for capture

The exhaust gas compositions of the traditional PPs, including NG-based MCFC, are summarized in Table 2 [48,50,76–82].

Although the overall exhaust gas compositions of the stand-alone MCFCs can vary significantly according to the fuels and operating conditions, the overall CO_2 composition of commercialized MCFC is known to be similar to that of NGCC. On the other hand, other gas compositions are slightly different from NGCC, as listed in Table 2. However, MCFC exit gas is emitted separately from the anode and cathode.

The CO_2 composition of the anode exit gas is 10 fold greater than that of the cathode because CH_4 reforming is conducted in the anode and CO_2 is accumulated in the anode during the operation. Furthermore, some fuel remains necessarily unreacted in the anode outlet gas because an excessive concentration loss of fuel should be avoided in the anode end region. Therefore, MCFC is generally operated with a fuel utilization range of 40–90%, which results in the anode outlet gas containing H_2, CH_4, CO, CO_2 and steam. Finally, these gases should be burnt and the generated heat used in the system, which increases the water and CO_2 concentration in the anode section to about 75% [34]. Therefore, CO_2 capture in the anode section can be more effective than in the cathode section.

MCFCs can be effectively utilized as the CO_2 separator by integration with the traditional thermal PP (PP-MCFCs). This is substantially different from the MCFC–GT hybrid system which primarily focuses on high electrical efficiency. PP-MCFCs can be operated with high energy efficiency while maintaining effective separation (or concentration) of CO_2 emitted from the PP. The key point which influences PP-MCFCs performance is the system layout and operating condition to maximize the heat utilization. Furthermore, MCFCs can be easily and effectively retrofitted to traditional capture processes such as absorption using MEA (mono-methanol-amine) and Selexol solvent as well as condensation units.

Therefore, extensive research using MCFCs to reduce CO_2 emissions has been carried out in power generation applications. Current research is focused on the following three different systems that are analyzed in the next sections: system using PP-emitted CO_2, system using CO_2 in the anode exit gas, and system integrated with coal gasification. Their technologies are reviewed in the next section, which describes the detailed results of the works.

2.2.1. System using CO_2 emitted from power plant (PP) as the reactant in the cathode

MCFCs in PP-MCFCs can act either as a CO_2 separator or a concentrator to capture CO_2 emitted in the PP. Although many PP-MCFCs processes have been proposed with various system layouts, the principle configuration is shown in Fig. 3.

In the system, the plant exhaust gas (or added air) is used as the reactant in the cathode and the depleted CO_2 gas is vented out to the atmosphere. On the other hand, CO_2 is captured from the anode exit gases after burning with air (or O_2).

Many works have reported the performance and CO_2 emission reduction of their systems. The results are summarized in Table 3 and technology analysis is described below.

In 2002, Campanari [80] investigated the energy efficiency and CO_2 capture capacity of the modern super critical (MSC) CFPP–MCFC hybrid system in which air added to the coal combustion exhaust gas of CFPP is fed to the cathode of MCFC using NG as the fuel, as shown in Fig. 4.

The cathode outlet gas is vented out to the atmosphere, whereas the anode outlet gas is supplied to a shift reactor which converts the gas into CO_2, H_2, and H_2O with compositions of 54.42%, 10.24%, and 34.90%, respectively. These gases are discharged from the reactor and

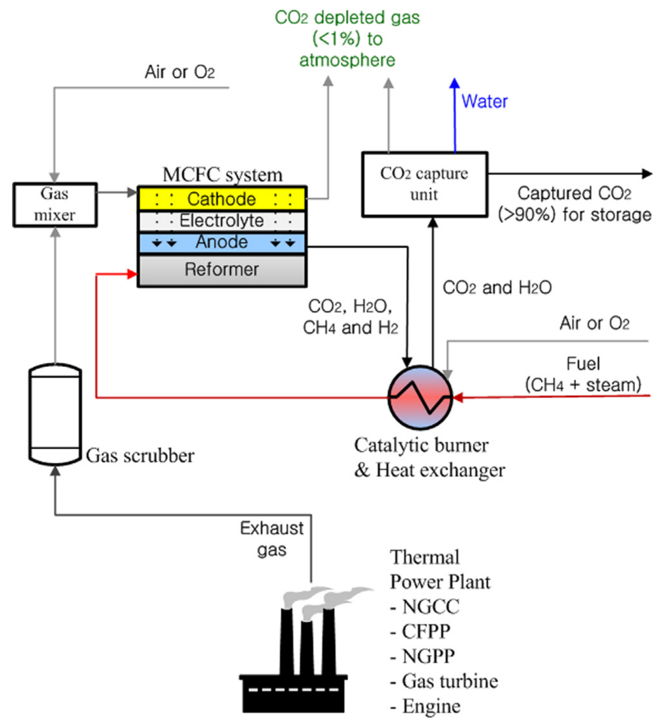


Fig. 3. Principle configuration of MCFCs integrated with a traditional thermal power plant (PP) for CO_2 capture.

H_2O is condensed, which increases the composition of CO_2 and H_2 to 83.16% and 15.66%, respectively. Subsequently, CO_2 is solely captured by physical absorption and H_2 is recycled in a steam cycle to be burnt for steam generation. According to the paper, the net energy efficiency of the system is similar to that of traditional MSC-CFPP and the calculated energy required to capture CO_2 is 52.9 MW. However, the system can significantly reduce CO_2 emissions, as listed in Table 3.

Sugiura et al. [76] also claimed that MCFCs can substantially reduce CO_2 emissions in an NGPP or NGCC. However, the authors did not describe the additional benefits of MCFC integrated with PP. Their system is shown in Fig. 5.

The authors highlighted that the stable operation of MCFC is a prerequisite to apply MCFC to the CO_2 separator in the process. Subsequently, they investigated many variables influencing the system performance and finally concluded that the P_{O2}/P_{CO2} ratio is the most crucial factor: performance improved with increasing ratio. The CO_2 and O_2 composition in the exhaust gas of the traditional NGCC is 4 and 13 mol%, respectively. Therefore, P_{O2}/P_{CO2} is 3.25, which is sufficiently higher than the theoretical value of 0.5 to maintain stable MCFC operation. Therefore, the authors claimed that the CO_2 composition in the NGCC exhaust gas can be readily decreased to 0.4% by MCFCs. On the other hand, the P_{O2}/P_{CO2} ratio in traditional NGPP is lower, at approximately 0.37, than that of the theoretical value for CO_2 capture. Therefore, O_2 should be added to increase the ratio and the authors claimed that a 20% makeup of air is optimum. According to this paper, MCFC can reduce the CO_2 concentration in the NGPP exhaust gas from 8% to 4.2% in the air-make-up condition. In addition, the authors claimed that MCFCs was operated with a current density of 100 mA/cm^2 and CO_2 utilization of 40% to obtain the CO_2 separation effect of MCFCs. However, they did not explain the separated CO_2 treatment in their paper.

In 2004, Lusardi [27] conducted a detailed analysis of MCFCs equipped with NGCC, as shown in Fig. 6.

GT exhaust gases are directly fed to the cathode and finally discharged from MCFCs. The heat in the cathode outlet gas is

Table 3
Performance and CO_2 emission reduction of PP-MCFCs reported in the literature.

Reference (year)	Systems (capacity; MW)	Efficiency (%)	CO_2 capture ratio (%)	CEC (kg of CO_2/kW h)	Captured CO_2 resources (CO_2 capture option)	Resource of CO_2 emitted to atmosphere
[80] (2002)	CFPP (450)–MCFC (198)	45.77 (net)	76.9	0.148	Anode outlet gas (physical absorption)	Some of CFPP exhaust gas
[76] (2003)	NGPP (600)–MCFC (94.5)	–	47.5	–	–	Some of NGPP or NGCC exhaust gas
	NGCC (600)–MCFC (94.5)	–	90			
[26] (2004)	NGCC (GT: 4.6)–MCFC (2)	–	53	0.229	Anode outlet gas (condensation	Some of NGCC GT exhaust gas
[83] (2004)	NGCC (GT: 4.6)–MCFC (1.6)	–	50%	–	Anode outlet gas (condensation	Some of NGCC GT and some of burnt anode exhaust gas
[50] (2009)	CFPP–MCFC	40 (overall)	39	0.253	Anode outlet gas (condensation	Some of CFPP exhaust gas
[1] (2010)	NGCC (GT: 271, ST:191)–MCFC (89)	58 (power efficiency)	80	0.071	Anode outlet gas burnt with O_2 (condensation)	Some of NGCC GT exhaust gas
[36] (2011)	NGCC (GT: 275 ST:161)–MCFC (102)	59 (power efficiency)	76	0.083	Anode outlet gas burnt with O_2 (condensation and cryogenic)	Some of NGCC GT exhaust gas
[35] (2013)	NGCC (800)–MCFC (212)	52.5	58.1	0.149	Anode outlet gas (membrane)	Some of NGCC exhaust gas
[37] (2013)	Reciprocating engine (6)–MCFC (1.8)	41	76	0.121	Anode outlet gas burnt with O_2 (compression)	Some of reciprocating engine exhaust gas

recovered in the steam generator and thereafter the gas is vented out to the atmosphere. Meanwhile, the anode outlet gas is burnt with air or O_2 in a burner and the heat generated in the combustion is transferred to the cathode outlet gas going to the steam generator. As a result, CO_2 in the anode outlet burnt gas is condensed for capture. Therefore, the only CO_2 emitted to the atmosphere is in the cathode outlet gas, which is only 32% of that of traditional NGPP (0.713 kg CO_2/kW h). Therefore, the authors claimed that the CEC of the system is below the standard CEC level recommended by IPCC for NGCC (0.286 kg CO_2/kW h). In addition, they reported that the system can capture 39.8 kmol/h of CO_2, which is 53% of CO_2 emitted from GT in NGCC. Furthermore, if the O_2 is used for anode outlet gas combustion, the CO_2 is concentrated to a final composition of 99%, which significantly enhances the capture ratio. In the system, absorption using MEA is assumed to be used as another capture option. The authors also highlighted two important operating conditions of MCFCs to separate CO_2 in the system: the CO_2 utilization factor should be maintained higher than 55% to avoid diffusion limited operating conditions and the CO_2 concentration should be kept higher than 5–6% to avoid side reactions.

In 2004, Amorelli et al. [83] analyzed the performance of the NGCC–MCFC system and its CO_2 capture capacity. The GT outlet gas of NGCC, with a CO_2 composition of 3 mol%, is directly fed to the cathode inlet, as shown in Fig. 7.

According to the paper, half of the CO_2 in the GT outlet gas is transferred to the anode to operate MCFC and the other half is vented out to the atmosphere from the cathode. On the other hand, CO_2 coming from the cathode is added to the anode inlet gas generated from the reformer, which increases the wet-based CO_2 composition in the anode from 25 to 55 wt%, and discharged from the anode. The anode outlet gas is primarily composed of steam, with trace amounts of CH_4, H_2 and CO. Steam is firstly eliminated by condensation and CO_2 is subsequently captured. The remaining gases are mixed with the cathode outlet gas and burnt in the catalytic burner. The heat released from the combustion is used for reforming and the cooled combustion gas is emitted to the atmosphere at a rate of 2.0 t CO_2/h. The authors claimed that this is 40% of traditional NGCC emissions and considering the 1.6 MW of power generated by MCFC, the CEC is estimated to be half that of general NGCC emissions. However, they reported that the pollutants remaining in the NGCC exhaust gases such as NO_x and SO_x can substantially deteriorate the system performance.

Milewski et al. [50] claimed that MCFCs integrated with CFPP can substantially reduce CO_2 emissions. The exhaust gas of CFPP is used in the cathode gas and the layout is shown in Fig. 8.

The anode outlet gases of MCFC sub-system using NG as the fuel are burnt with O_2 in a catalytic burner. The combustion gas is cooled to produce water and separate the CO_2 after using their heat to increase the temperature of steam and CH_4 for reforming as well as the CFPP exhaust gas going to the cathode inlet. In addition, the heat to increase the temperature of cool water from steam turbine (ST) is obtained from this combustion gas. Subsequently, the heated water goes to the boiler at a temperature of 220 °C after obtaining additional heat from the cathode outlet gas in the heat exchanger. Air is added to the CFPP outlet gas with CO_2 and O_2 composition of 15 and 5.98 mol%, respectively. The temperature of these gases is increased from 120 °C to 338 °C by the heat released from the anode outlet gas combustion. Thereafter, the gases are fed to the cathode inlet and finally emitted to the atmosphere with a CO_2 composition of 8.5 mol%. According to the paper, the energy efficiency and CEC of the system are 10% and 22.3% higher than those of traditional CFPP, respectively.

In 2010, combining the advantageous features of MCFCs previously described, Campanari et al. [1] introduced their NGCC–MCFC system integrated with a CO_2 capture unit and claimed that

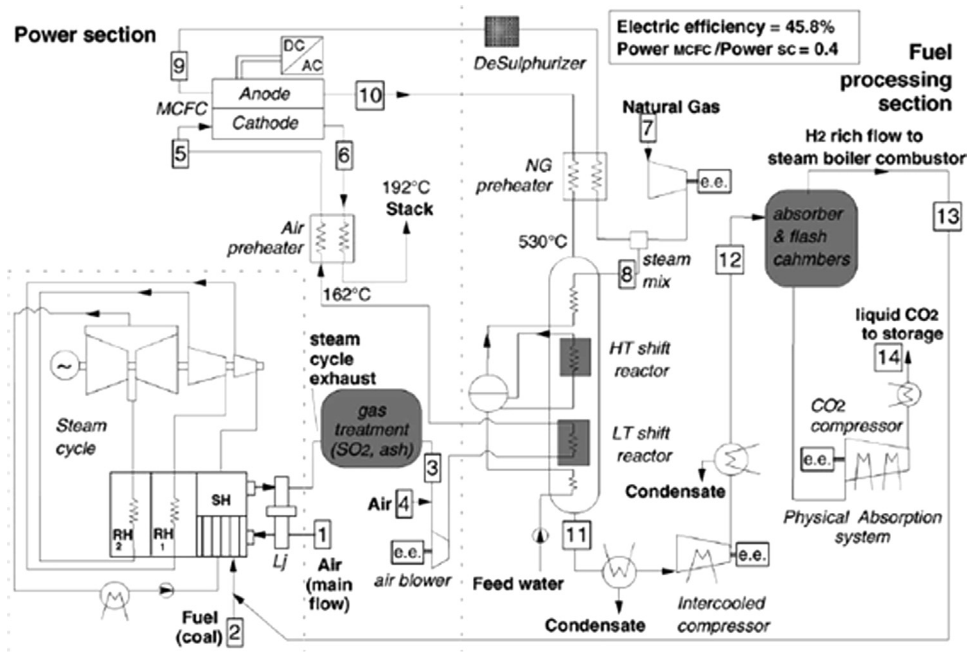


Fig. 4. CO_2 separation from a conventional power plant (PP) by integration with a MCFC plant [80].

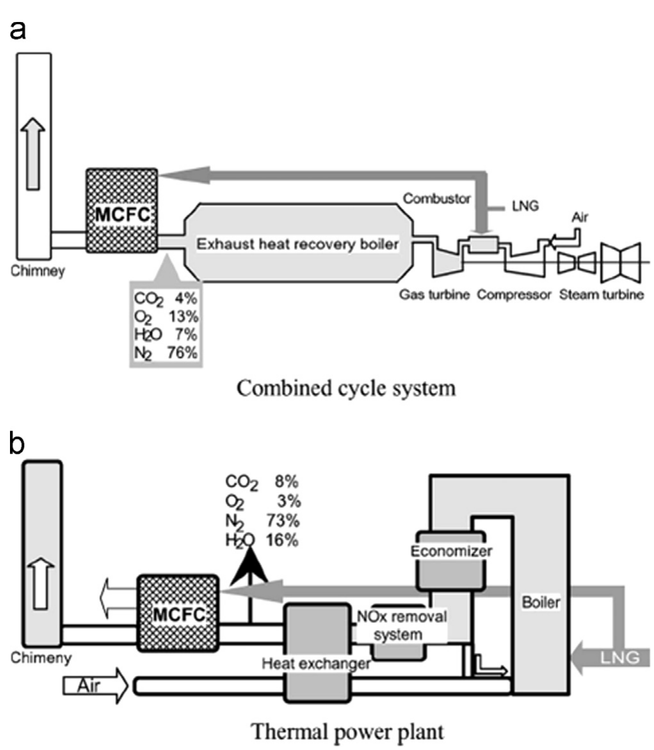


Fig. 5. Schematic diagram of NG-based thermal power plants (PPs) with MCFC [76].

it exhibited very high capture efficiency. The process layout is shown in Fig. 9.

The process is designed to maximize the synergy effects between NGCC and MCFC. CO_2-depleted cathode outlet gases are emitted to the atmosphere after their sensible heat is transferred to the reformer and heat recovery steam generator (HRSG). On the other hand, the anode outlet gas is mixed with O_2 to complete the combustion of the trace amount of remaining unreacted fuel. Very rich CO_2 (41.6 mol%) in this combustion gas was captured by condensation and liquefied to 25 °C at 110 bar. After analyzing the system performance according to various conditions and assumptions, the authors claimed that the net electric efficiency was almost the same as that of their referenced NGCC. In addition, the CO_2 capture efficiency was about 80% and the energy consumption for CO_2 capture was calculated to be 0.82 MJ/kg CO_2. When using absorption with MEA in traditional NGCC, the CO_2 capture efficiency is known to be about 80%, which results in an energy consumption of 3.20 MJ/kg CO_2. Therefore, the authors claimed that their process can save 75% of the energy for CO_2 capture compared to conventional systems. They also reported that the system performance is not sensitive to the variation of many factors including the current density, CO_2 and fuel utilization, steam to carbon ratio and CO_2 composition in the cathode inlet gas. In addition, if some of the cathode outlet gas passing the HRSG is recycled to the cathode via the GT, the authors claimed the traditional GT could be used in the system [84], which can eliminate the effort to design a new GT in the MCFC–GT system.

In 2011, the same authors [36] investigated a similar NGCC–MCFC system equipped with cryogenic separation for CO_2 capture and reported its performance in comparison to the above work. In the paper, the finalized anode exhaust stream is cooled in the HRSG and sent to a cryogenic CO_2 removal section to be liquefied; the remaining gas is recycled to GT for re-combustion. According to their simulated results, the system has the potential to achieve a CO_2 capture ratio of 76% maintaining its electric efficiency at 1% higher than that of previous work, while the CEC of the work was slightly larger than that of the previous system.

In 2013, Greppi et al. [35] proposed the installation of gas separation membranes (GSM) to NGCC–MCFCs to finally separate enriched CO_2 emitted from MCFCs as shown in Fig. 10.

The exhaust gas from the GT of NGCC (S09 in Fig. 10) is fed to the cathode with a CO_2 composition of 4.2% and it decreases due to cell reaction. CO_2-depleted cathode outlet gas (S11) is sent to the ST to supply its heat to generate steam. On the other hand, CO_2-enriched anode outlet gas (S10) is condensed to separate some water (S14) from the gases including CO_2, CH_4, H_2 and CO. Subsequently, CO_2 in this stream is separated by GSM (S16) for capture and the other segregated stream (S15) is recycled to the anode (S04), steam reformer (S03) and GT. From the simulation, the authors claimed the performance of the system was substantially positive under the optimized conditions and the economy

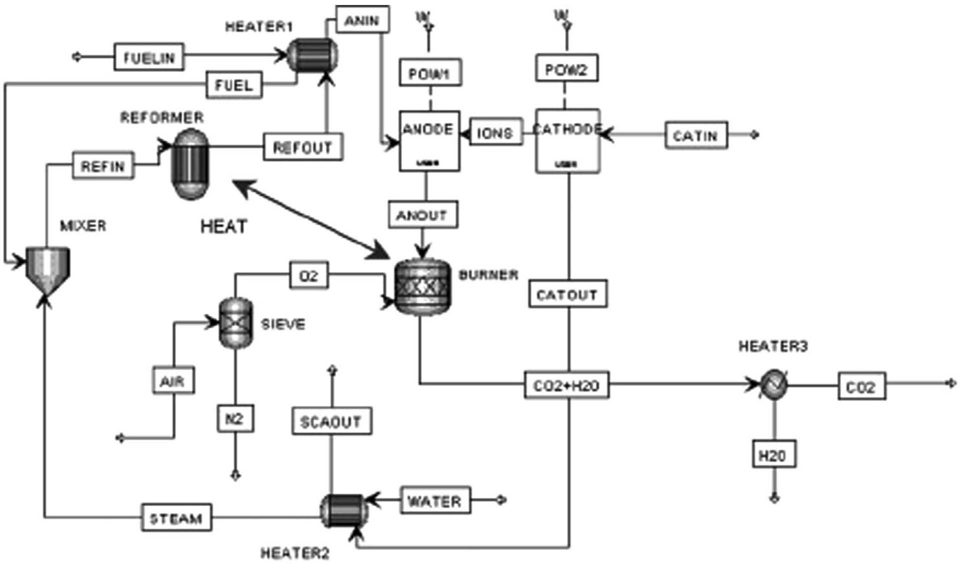


Fig. 6. Integration of GT, MCFC and an anodic catalytic burner for CO_2 separation: plant scheme for operation at 1 atm [26].

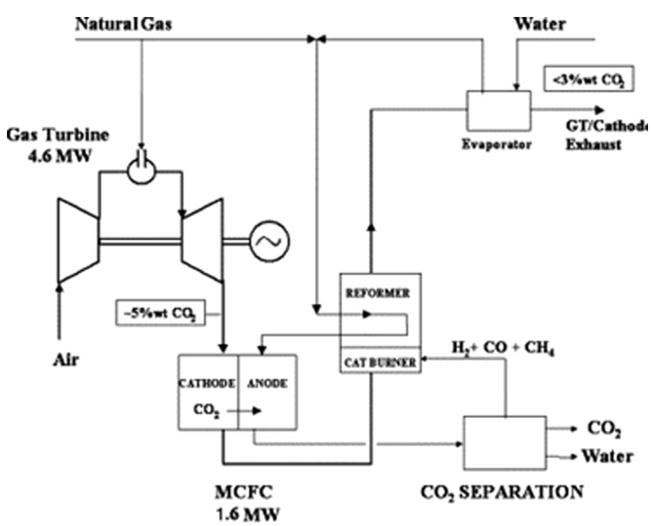


Fig. 7. Hybrid atmospheric pressure MCFC scheme for CO_2 capture [83].

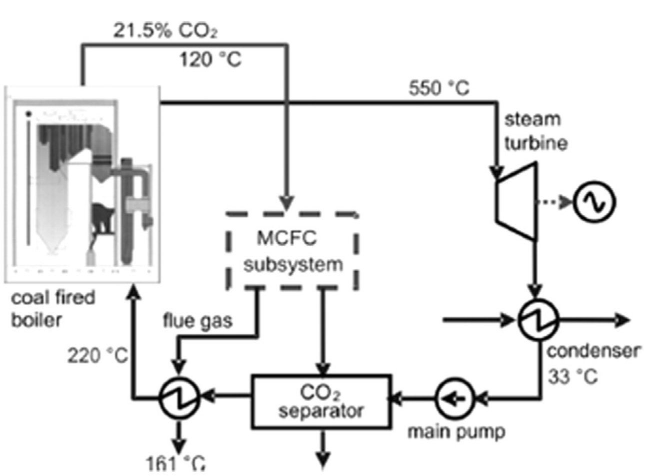


Fig. 8. Flow diagram of MCFCs integrated with CFPP [50].

was better than that of NGCC–MCFCs combined with the conventional amine absorption for capture in all scenarios. In addition, although the maximum hot-spot zone was created in MCFC and this is the main problem to limit the performance, the authors highlighted that it could be addressed by improving the flow distribution within the cell and recycling the cooled cathode flow.

In 2013, Sánchez et al. [37] analyzed the MCFCs integrated with an NG-based reciprocating engine as shown in Fig. 11.

The performance of the system including the electric efficiency and CO_2 emission reduction was compared to two other reference cases. One is a stand-alone power generation system solely using a reciprocating engine without capture and the other is the same system coupled with a MEA absorption process for capture. As a result, the energy consumption for absorption by MEA reduced the original efficiency of the stand-alone engine by 10%. However, the authors claimed that, although the CO_2 emission of their MCFC-engine hybrid system was 2.5 fold larger than that of a MEA coupled system, it can be operated with similar efficiency to a stand-alone engine because the power consumption for CO_2 capture and compression as well as parasitic energy losses can be minimally maintained in the system.

2.2.2. System using CO_2 in the anode exit gas as the reactant in the cathode

MCFCs can also be used as an effective CO_2 separator or concentrator by itself. Slight amounts of H_2 and CH_4 in the anode exit gas are burnt in a catalytic burner with air (or O_2), as shown in Fig. 12, and the generated heat can be diversely used, such as for steam generation and air heating.

Some CO_2-enriched burnt gas is recycled to the cathode and the remaining gas is captured (dotted arrow line in Fig. 12). Some studies have reported their system performance with CO_2 capture ratio and these are summarized in Table 4. In addition, their technology is reviewed below.

In 1997, Reimer et al. [85] were one of the first research groups that reported the effective application of MCFCs as a CO_2 separator in the power application field. Among the many processes they analyzed, the system with the best performance is shown in Fig. 13.

De-sulfurized NG and steam are directly fed to the anode as the fuel because their MCFCs is assumed to be the internal reforming system. The mixture of anode outlet burnt gases and air is supplied to the cathode as the reactant. The heat in the cathode outlet gas is recovered in the gas heater and the HRSG, which cools the gases. Thereafter, the stream is supplied to the CO_2 capture

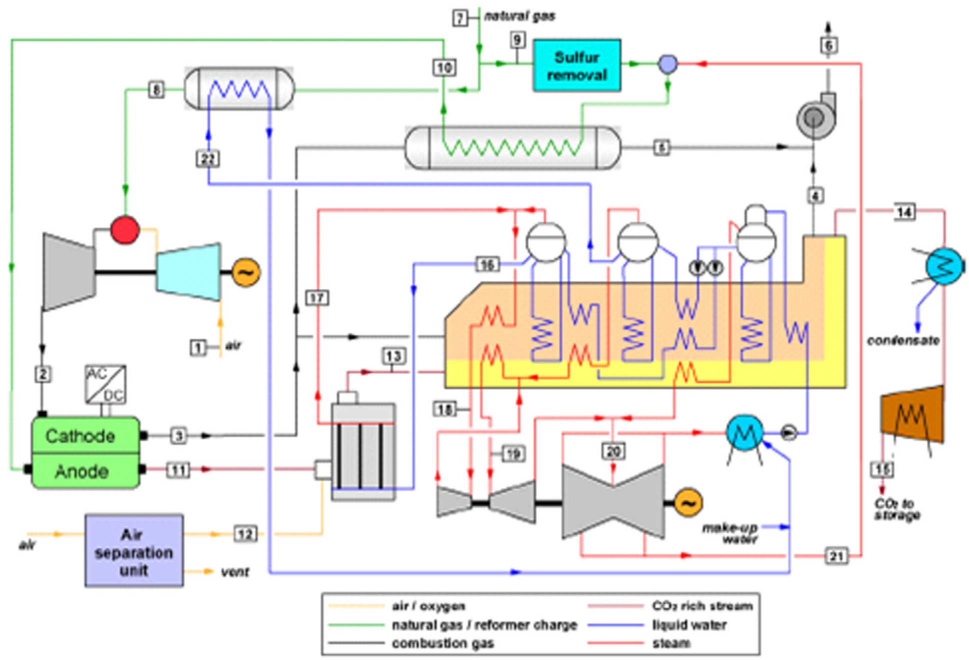


Fig. 9. Plant layout showing the integration of the MCFC in a combined cycle, with CO_2 separation after pure oxygen combustion of the cell anode exhaust [1].

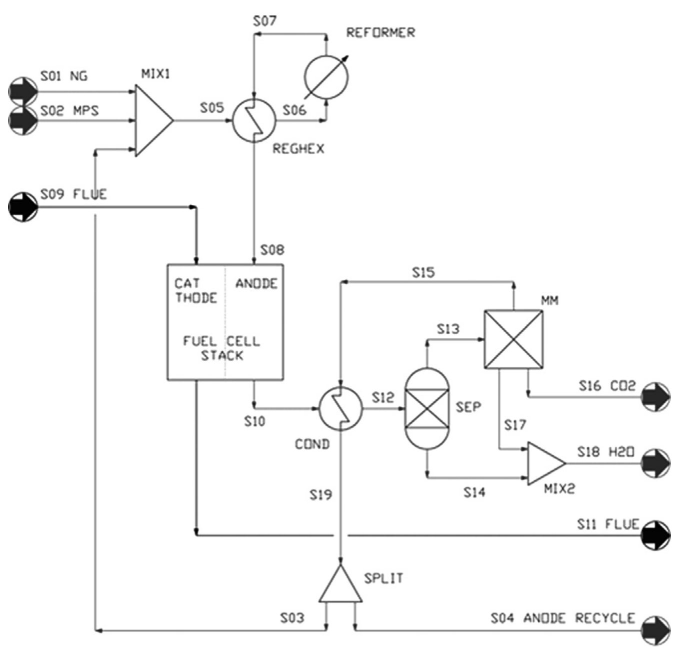


Fig. 10. Process flow diagram for the MCFC unit [35].

unit using MEA absorption. The recovered CO_2 is highly pressurized and transported via a pipe line. According to the paper, the performance and capture ratio were significantly high. This work also investigated the effects of many variables on the system performance such as CO_2 recycle ratio, pre-combustion, and reforming option.

Akai et al. [86] presented the CO_2 capture capacity of the MCFC–ST hybrid system equipped with the capture process. Coal-derived NG was used as the fuel in the MCFC. The capture process uses two chemical absorption options employing K_2CO_3 and MEA as the solvent. The system configuration is shown in Fig. 14.

CH_4 extracted from coal gasification and O_2 are fed to the anode. After passing the heat exchanger, the anode outlet gas is divided into two lines: one is recycled to the anode and the other is fed to a K_2CO_3 scrubber for CO_2 elimination. The CO_2-depleted outlet gas from the absorber diverges again into two streams: one line goes to ST for power generation after passing the heat exchanger to be finally condensed, and the other line is provided to the cathode after burning with air coming from the air compressor. The cathode outlet gas is passed through GT, HRSG and MEA scrubber for capture and finally emitted to the atmosphere. The authors calculated a system efficiency of 53.1% without any CO_2 capture unit, but this was lowered by 10% due to CO_2 capture in the system. Instead, the authors claimed a potentially very high CO_2 capture ratio for the process.

Lorenze and Fragiacomo [87] analyzed the performance and CO_2 capture capacity of the MCFC–GT hybrid system, as shown in Fig. 15.

The authors claimed that many innovative options had been considered to maximize the effect of MCFCs on CO_2 capture. One of the anode outlet gases is mixed with dry air and CH_4 is burnt in the burner (B1 in Fig. 15). The heat of this combustion gas is supplied to the heat exchanger and the gas is then fed to the cathode together with O_2 and emitted from the cathode. The heat in the cathode outlet gas is used in HRSG, and the gases are thereafter emitted to the atmosphere. The generated steam is used for reforming or other applications. Another stream of the anode outlet gas is converted to enriched CO_2 and steam by combustion with air in another burner (B2 in Fig. 15). Thereafter, CO_2 in the gas is captured by condensation with a composition of 98% and 99%. The heat generated from the condensation is transferred to increase the temperature of the air fed to drive GT. Therefore, the process is considered to be an indirect MCFC–GT hybrid system and its feature is to maximize the potential heat utilization supplied by the system. The sensible heat of the cathode outlet gas is the primary source required for HRSG and the heat in the outlet gas from the micro GT (MGT) and from B2 is used to generate steam. Therefore, the authors claimed a very high system performance and a CEC value of only 1/3 of the traditional NGCC system.

2.2.3. System integrated with coal gasification and using waste heat

In addition to the two main options described above, MCFCs can be very diversely used to reduce CO_2 emissions in the plant. Two such cases are presented below.

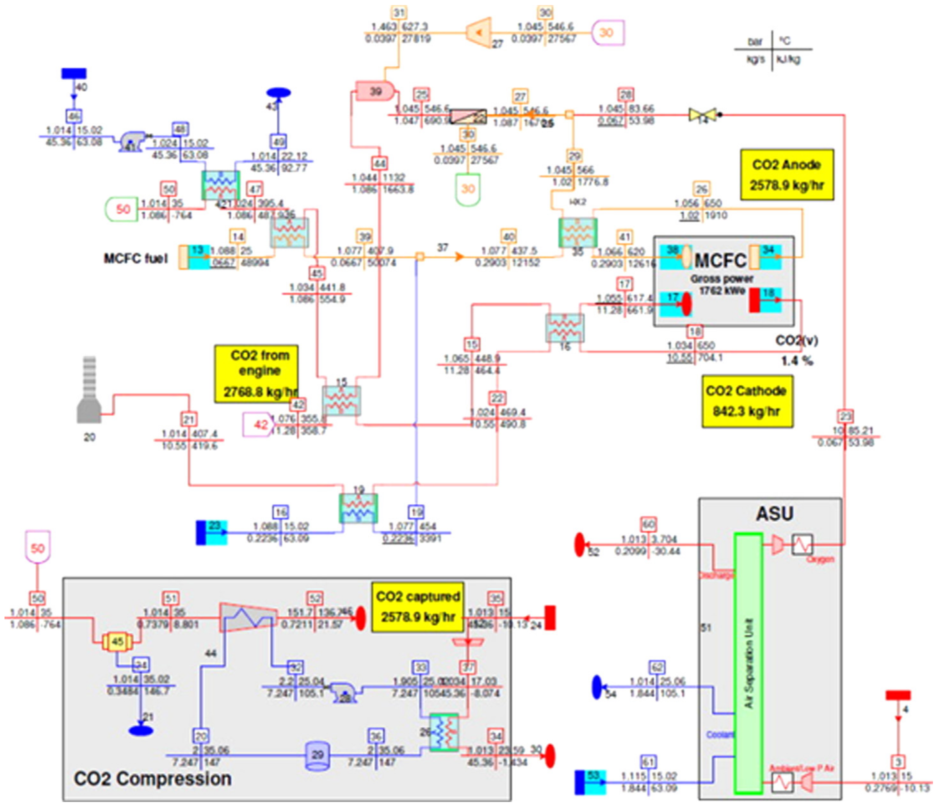


Fig. 11. MCFC carbon capture integration layout [37].

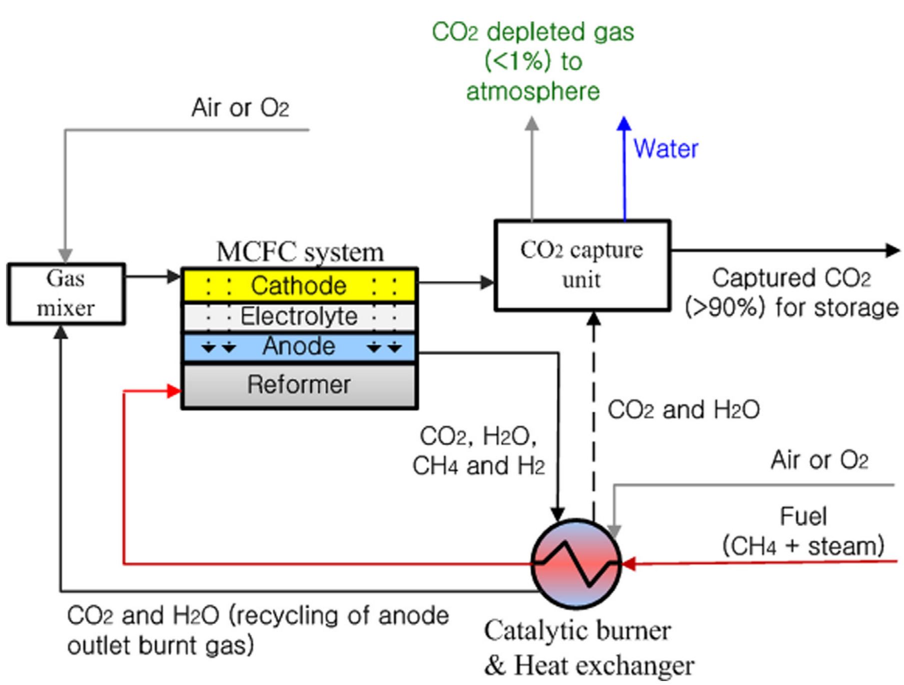


Fig. 12. MCFCs operation using CO_2 in the anode outlet gas as the reactant in the cathode for CO_2 capture.

Wang et al. [78] investigated the performance of the MCFCs integrated with the coal gasification process known as absorption-enhanced reforming (AER) which produces H_2 and $CaCO_3$ from lignite coal and steam, as shown in Fig. 16.

The produced H_2 is used in MCFC and CaO is employed for CO_2 (and sulfur) capture to generate $CaCO_3$. CaO is recovered by removing the CO_2 in the sorbent regeneration unit and recycled to AER process. According to the paper, the performance of MCFC in the system is similar to that of traditional NG-based MCFC and the CEC of the system is very high. The authors attributed this to the use of coal with a high carbon composition of 73.9% as the fuel and the high energy consumption required to gasify the coal. They claimed, however, that CO_2 can be very effectively captured at a high composition of 64% and balance steam. Furthermore, the

Table 4
Performance and features of the systems using MCFCs to reduce CO_2 emissions reported in the literature.

Reference (year)	Systems (capacity; MW)	Efficiency (%)	CO_2 capture ratio (%)	CEC (kg of CO_2/kW h)	Captured CO_2 resources (CO_2 capture option)	Resource of CO_2 emitted to atmosphere
[85] (1997)	MCFC ($100 \times 5 = 500$)–ST (68)	55.5	92	–	Anode outlet gas burnt with air (chemical absorption using MEA)	Cathode outlet gas
[86] (1997)	IG–MCFC–ST	43.5	68–79	–	Anode outlet gas (of two divided streams) burnt with air (chemical absorption using K_2CO_3, MEA)	Cathode outlet gas
[87] (2010)	DIR-MCFC[a] (0.5)–MGT (0.1)–ST	0.52	–	0.101	Anode outlet gas (of two divided streams) burnt with air (condensation)	Cathode outlet gas
[78] (2006)	IG–MCFC (0.2)	44.7	–	0.751	Coal (Ca looping)	Coal
[88] (2009)	MCFC (2.4)–SGT (0.33)	48.2	583 t CO_2/y	0.751	CH_4 (condensation)	MCFC outlet gas

[a] Direct internal reforming MCFC.

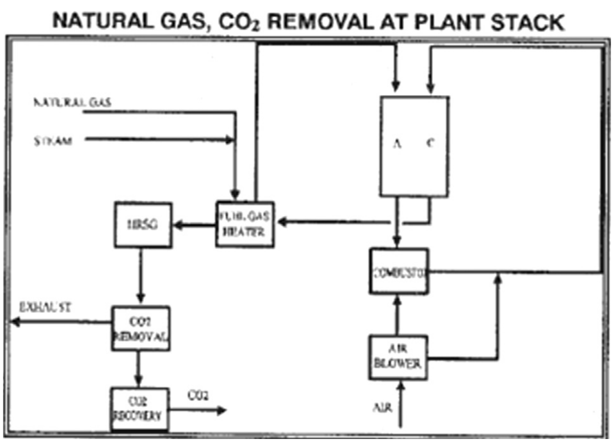


Fig. 13. Process flow diagram of MCFCs using the anode outlet burnt gas in the cathode for CO_2 capture [85].

authors claimed that the system can be considered a zero-emission process due to the absence of any emitted NO_x and SO_x in the process.

Pak et al. [88] analyzed the economy of their proposed system consisting of commercial MCFC integrated with a conceptual steam GT (SGT) system. Their key point is the heat utilization of the MCFC outlet gas, as shown in Fig. 17.

The efficiency and outlet gas temperature of the MCFC assumed in the paper were 47% and 340 °C, respectively. The MCFC outlet gas is vented to the atmosphere after delivering its sensible heat to the returning water in HRSG. The steam generated from HRSG is mixed with O_2 and fuel and this gas mixture is then burnt to generate CO_2 and steam under very high pressure with a temperature of 1000 °C, which drive SGT. Subsequently, CO_2 is separated from the steam by condensation. Finally, the separated CO_2 is liquefied under high pressure. The paper reported a system performance slightly higher than that of commercial MCFC system, as listed in Table 4. However, the economy of the system is low due to the high energy consumption required for CO_2 capture. However, the authors claimed economically viable system operation at a CER price consistently over 20 USD/t CO_2.

3. Challenges

3.1. Full reliability of the operation of stand-alone MCFCs

Although MCFCs can be effectively used as the CO_2 separator and concentrator for CO_2 capture, many problems require solution before it can enter practical use. Firstly, MCFCs should be successfully commercialized as the distributed power generation. This can be realized by the further stable operation of stand-alone MCFCs with full reliability. Although MCFCs has been actively installed in Korea, in fact many problems are still being reported in terms of system operation. MCFCs technology has greatly developed to the level of commercialization due to great research efforts. The overall life span of MCFCs is reported to be 5 years. Anode and cathode have been used for 3 years or more without serious degradation. However, some crucial issues remain to be addressed for further stabilized operation of the system, which includes matrix drying, cathode dissolution and Ni precipitation and cathode current collector corrosion [58,89].

The production cost of MCFCs is currently reported at under 3000 USD per kW and this reduced to 60% since the first commercialization was installed in 2003. The cost of electricity generated by MCFCs is known to be 0.15 USD per kW h in the U.S. and this value is already competitive with grid parity in some markets [89]. Nevertheless, the system cost should be further reduced for market extension. Therefore, the power of the MCFCs module should be at least 10-fold larger [38] than that of the relatively small scale (> 2 MW) current installation and this is also a very significant factor in applying MCFCs to CO_2 capture equipment.

The other challenges in terms of MCFCs as the CO_2 separator and concentrator for capture are summarized below.

3.2. Impurities in PP exhaust gas

Impurities in the PP exhaust gas can significantly damage the MCFCs. The molten carbonate salts such as K_2CO_3/Li_2CO_3 used as the electrolyte of MCFCs are very susceptible to contamination by the H_2S, SO_x and NO_x in the exhaust gas forming sulfates and nitrates. Fine particles in the exhaust gas block the active sites (or pores) of the electro-catalyst and mercury, even at trace amounts, deactivates the electro-catalyst. Therefore, such impurities in the exhaust gas must be reduced under the tolerable level of MCFCs, as has been analyzed in detail in the literature [88,90–94].

Regarding steam in PP, although the detailed mechanism has not yet been completely elucidated, it is known to be useful to accelerate the CO_2 soluble rate into ${CO_3}^{-2}$ in the cathode [76].

3.3. Temperature control of PP exhaust gas and hot-spots in fuel cells

Compared to the traditional PP exhaust gas temperature of approximately 120 °C that for reaction in MCFCs should be approximately 650 °C. However, such a high temperature is difficult to simply achieve and the gas cannot be cleaned at this temperature. In addition, the connection of PP and MCFCs may be very difficult. Therefore, an innovative system design is required

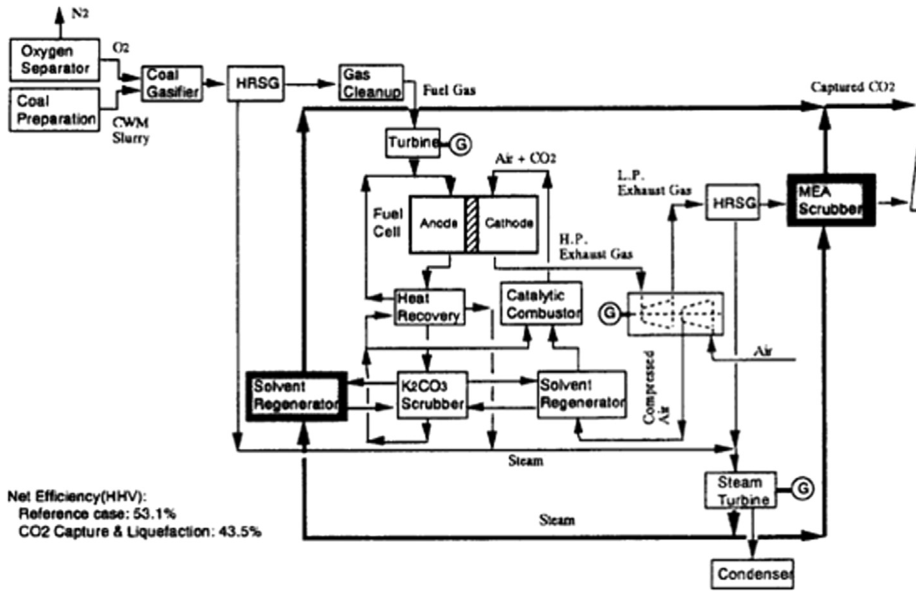


Fig. 14. Process flow diagram for the MCFC system with CO_2 capture [86].

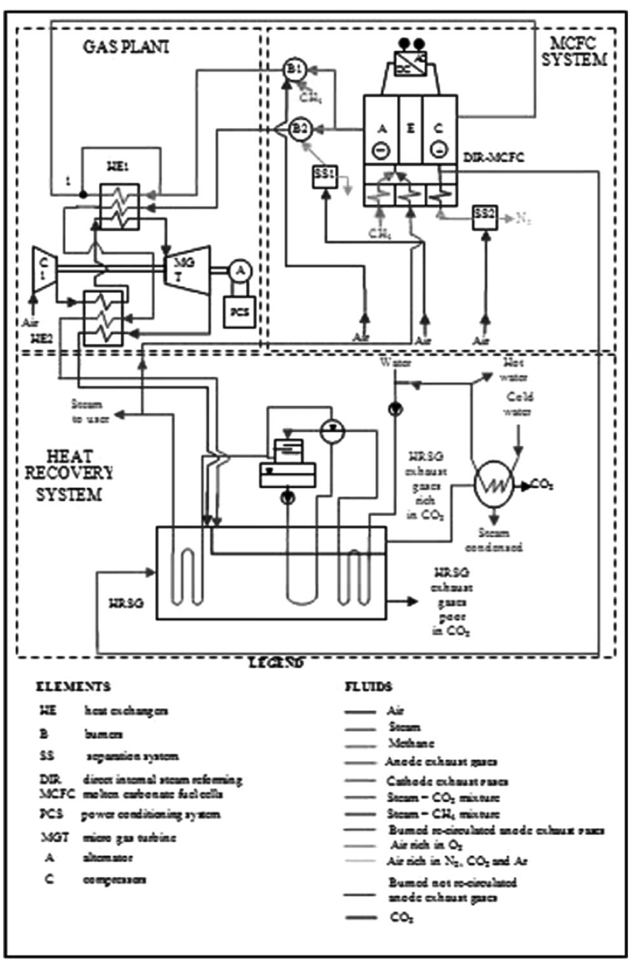


Fig. 15. Technical layout of the eco-friendly hybrid plant with MGT and MCFC system [87].

for diverse utilization of the heat released from the system and novel equipment should be developed to effectively connect the two systems. The maximum hot-spot temperature can partially occur within the cell because many different temperature streams are substantially recycled within the system in a complicated manner, which may be main process limitation. However, the solutions to this issue are presented in the paper [35].

3.4. *The P_{O2}/P_{CO2} ratio in the cathode and combustion with pure oxygen*

One of the most significant factors to influence the capture performance is the P_{O2}/P_{CO2} ratio in the cathode. Although it increases the performance in proportion to its value, the optimum value should be found at a given CO_2 concentration. The inherent NGCC outlet gas can be directly used as MCFC cathode gas because the P_{O2}/P_{CO2} ratio is maintained at over 0.5, which is the theoretically minimum value for cell reaction. However, the outlet gas composition of other PP such as NGPP, CFPP and IGPP should be controlled by the other gas stream within the system, which substantially complicates the management of the gas stream to maintain the optimum P_{O2}/P_{CO2} ratio.

Combustion of the anode exit gas is almost mandatory in the system. Many papers have reported the very effective use of O_2 for the combustion rather than air because it reduces the unnecessary N_2 feeding and further enriches the CO_2 composition. However, O_2 production remains very energy intensive, which severely restricts the viability of this CO_2 capture process. Currently, the cryogenic air separation unit is most widely used to separate O_2 from the air. It consumes energy in the ranges of 250–350 kW h/t of pure O_2 [95–97]. Therefore, the main problem is whether the energy consumption is worthwhile in improving the CO_2 capture ratio in the system. Many papers employing O_2 combustion have not addressed this issue at all and not clearly commented on its alternatives. If the economics and technology for O_2 production are improved, almost all issues regarding CO_2 capture will be addressed.

4. Conclusions

This review on the potential contribution of MCFCs to CO_2 emission reduction in power plants examined the very latest

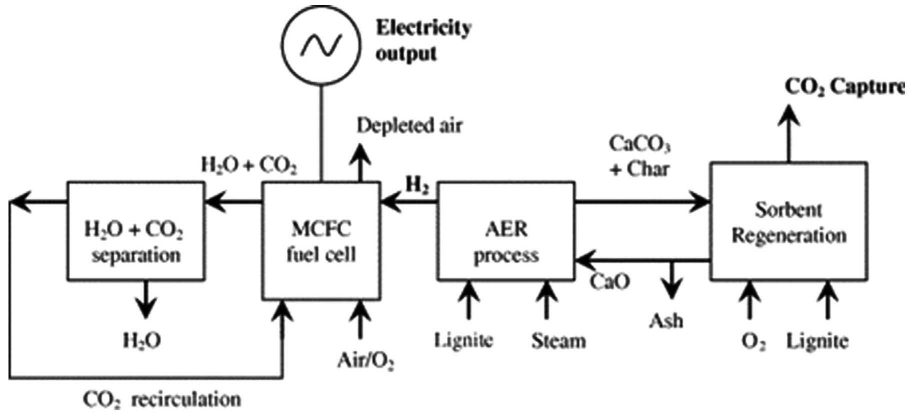


Fig. 16. Combination system of AER with MCFC [78].

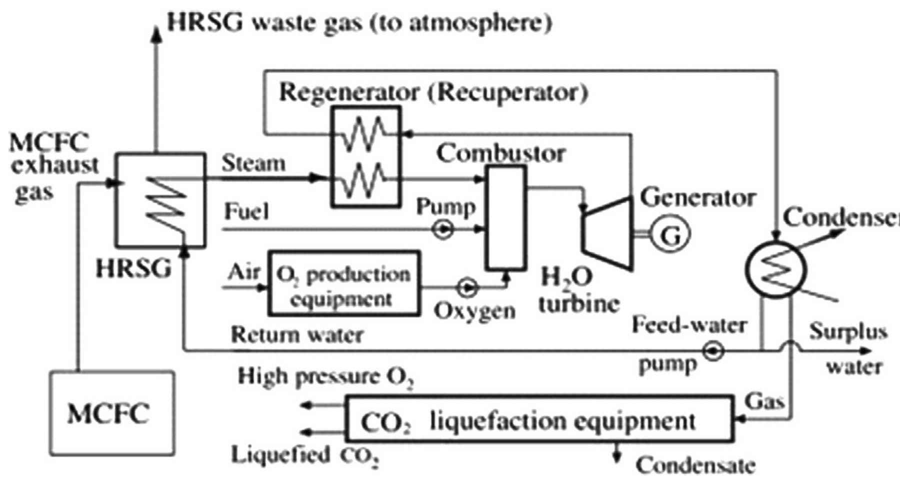


Fig. 17. Schematic structure of the proposed CO_2 capturing power generation system [88].

research works, from which the following conclusions can be derived.

– Considering the heat recovery, the overall efficiency of stand-alone MCFCs can be increased to approximately 75% and the CO_2 emissions reduced at least under 236 kg/MW h. In addition, the operating efficiency of the MCFC–GT hybrid system will be further increased in the future.
– MCFCs can act as a CO_2 separator or concentrator when integrated with the traditional thermal power plants. In the various systems analyzed, the CO_2 emitted in the plant can be effectively concentrated for capture without reducing the system efficiency.
– MCFCs can also be used as an effective CO_2 separator or concentrator by itself. A small amount of fuel remaining in the anode outlet gas is burnt to produce additional CO_2 and heat. The CO_2 enriched in the burnt gas can be effectively captured and used as a reactant in the cathode by recycling. The heat released can be diversely used in the system.
– Many technologies can be used to capture the CO_2 that is separated and concentrated by MCFCs. These include commercialized absorption processes using Selexol and MEA as the solvent and condensation as well as the future technologies such as membrane, absorption with K_2CO_3 based solution and Ca-looping. However, condensation seems to be the most advantageous due to its relative simplicity and ease of application to the heat management in the system.
– Although MCFCs has great potential to reduce CO_2 emissions in power plants, its present application has remained very limited by its high cost and small scale operation. However, the MCFCs will substantially contribute to the reduction of CO_2 emissions in power generation applications in the near future.

Acknowledgments

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